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L. Mathieu, D. Heuer, R. Brissot, C. Le Brun, E. Liatard, J.-M. Loiseaux,
O. Méplan, E. Merle-Lucotte, A. Nuttin and J. Wilson,
Laboratoire de Physique Subatomique et de Cosmologie ,
53, avenue des Martyrs, F-38026 Grenoble Cedex, France

C. Garzenne, D. Lecarpentier, E.Walle,
EDF-R&D, Département SINETICS,
1 av du Général De Gaulle, 92140 Clamart, France

Abstract

A re-evaluation of the Molten Salt Breeder Reactor concept has revealed problems related to its safety and to the complexity of the reprocessing considered. A reflection is carried out anew in view of finding innovative solutions leading to the Thorium Molten Salt Reactor concept. Several main constraints are established and serve as guides to parametric evaluations. These then give an understanding of the influence of important core parameters on the reactor's operation. The aim of this paper is to discuss this vast research domain and to single out the Molten Salt Reactor configurations that deserve further evaluation.

Introduction

In order to reduce CO_2 emissions in the coming decades, and, as a result, to mitigate global warming, it appears necessary to stabilize or, better, to reduce the use of fossil fuels. Resorting to a sustainable version of nuclear power may help replace classical energy production partially and thus satisfy an increasing world energy demand while conserving the climate and natural resources. The Generation IV International Forum for the development of new nuclear energy systems [1] has established a set of goals as research directions for nuclear systems: enhanced safety and reliability, reduced waste generation, effective use of uranium or thorium ores, resistance to proliferation, improved economic competitiveness. Molten Salt Reactors (MSR) are one of the systems retained by Generation IV. MSRs are based on a liquid fuel, so that their technology is fundamentally different from the solid fuel technologies currently in use. Some of the advantages specific to MSRs (in terms of safety/reliability, for example) originate directly from this characteristic [2]. Furthermore, this type of reactor is particularly well adapted to the thorium fuel cycle (Th-^{233}U) which has the advantage of producing less minor actinides than the uranium-plutonium fuel cycle (^{238}U-^{239}Pu) [3, 4]. Moreover, while breeding or regeneration in the U-Pu cycle can be obtained only with a fast neutron spectrum, in the Th-^{233}U fuel cycle it can, in principle, be obtained with a more or less moderated neutron spectrum. In a thermal neutron spectrum, poisoning due to the Fission Products (FP) being worse than in a fast neutron spectrum, the rate at which fuel reprocessing is performed can become a major issue. Because, in an MSR, the fuel is liquid, continuous extraction of the FPs is a possibility. Although MSRs can be operated as incinerators they will be discussed in this paper only as electricity producing critical systems.

In 1964, the Molten Salt Reactor Experiment (MSRE) was initiated at the Oak Ridge National Laboratory (ORNL). Generating 8 MWth of power, the reactor was operated without problems and with different fuels (^{235}U then ^{233}U) over several years. The expertise gained during this experiment led, in the 1970s, to the elaboration of a power reactor project, the Molten Salt Breeder Reactor (MSBR [5]). The studies demonstrated that fuel regeneration is possible with the thorium fuel cycle in an epithermal spectrum, provided very efficient and, as a consequence, constraining, on-line chemical reprocessing of the salt is achieved. Over the past few years, the MSBR has been reassessed in the light of new calculating methods [3, 4] so as to elaborate a new reactor concept that we call the Thorium Molten Salt Reactor (TMSR).

The MSBR suffered from several major drawbacks and was discontinued. The goal being, at the time, to obtain as high a breeding ratio as possible, the on-line chemical reprocessing unit considered had to process the entire salt volume within 10 days and this was very complex [6]. Because of this complexity, the project is often considered unfeasible. In addition, recent calculations have shown that the global feedback coefficient for this system is slightly positive. This contradicts the results that had been presented. The difference is probably due to the fact that, at the time, the compositions were handled in a homogeneous way while they are handled heterogeneously today. This critical issue makes the MSBR a potentially unstable system in some situations.

The aim of this paper is to present solutions to these problems. In our search for better reactor configurations, we have identified several constraints that are discussed in the first part of this work. We then discuss the impact of the various reac-

tor parameters on these constraints, i.e. chemical reprocessing, channel size, fuel volume and the proportion of heavy nuclei (HN) in the salt. A synthesis of these studies is set forth in the last section.

This work is based on the coupling of a neutron transport code (MCNP [7]) with a materials evolution code. The former calculates the neutron flux and the reaction rates in all the cells while the latter solves the Bateman equations for the evolution of the materials composition in the cells. These calculations take into account the input parameters (power released, criticality level, chemistry,...), by adjusting the neutron flux or the materials composition of the core on a regular basis. Our calculations are based on a precise description of the geometry and consider several hundreds of nuclei with their interactions and radioactive decay; they allow fine interpretation of the results. All the data discussed in this paper result from the evolution of the reactor over 100 years.

1 Constraints

We identify five major constraints in this study: safety, chemical reprocessing feasibility, fuel regeneration capability, materials life span, and initial inventory. Other constraints could be considered, such as waste minimization, thermal-hydraulics, or proliferation resistance but we concentrate essentially on the above five major constraints. We seek to understand the impact of the reactor's defining parameters on these constraints. In so doing, we can single out the best reactor configurations according to the weight assigned to each of the constraints.

1.1 Safety

In the work we present here this constraint concerns essentially the evolution of the feedback coefficients that should be negative. The more kinetic aspects of the reactor's safety properties are not considered here. Additionally, the ways in which we change the concept do not modify the other MSR safety properties, such as fuel dumping and the fact that MSRs are free of high pressure areas.

The feedback coefficients, $\frac{dk}{dT}$, are a measure of the variation of the multiplication factor (dk) with the temperature of the core or of a portion of the core (dT). The global feedback coefficient can be broken up into several strongly uncorrelated partial coefficients, each of which characterizes the variation of a specific parameter: the effects due to the expansion of the salt[1], and the purely thermal effects of the salt and of the graphite. This reads:

$$\frac{dk}{dT}\Big)_{total} = \frac{dk}{dT}\Big)_{density} + \frac{dk}{dT}\Big)_{Doppler} + \frac{dk}{dT}\Big)_{graphite}$$

In order for the reactor to be intrinsically safe, a temperature increase must not induce an increase of the reactivity and, as a consequence, of the power released by fissions. For this reason, the $\frac{dk}{dT}\big)_{total}$ coefficient must be negative. The thermal kinetics of the graphite, which is heated by gamma radiation and cooled by the salt, is much slower than that of the salt. Making allowance for this delay between the heating of the salt and the heating of the graphite, the coefficient for the salt alone, that is the sum $\frac{dk}{dT}\big)_{density} + \frac{dk}{dT}\big)_{Doppler}$ must also be negative. The degree of safety can be further increased if the density coefficient is made negative. This implies that any local loss of density, e.g. because of a bubble, decreases the reactivity of the system.

The uncertainties on these values are related to statistical errors that are well identified and can be reduced, but also to systematic errors that are not quantified and are related, for example, to uncertainties in cross section evaluations. For this reason, the feedback coefficients must be sufficiently negative to ensure unambiguous stability.

1.2 Feasibility of the Chemical Reprocessing

The term feasibility reflects the complexity associated to the chemical reprocessing. Indeed, some of the separation processes are considered too difficult to be implemented. This can have several causes: the processes considered are not well understood or mastered, the flow of materials to be processed is too large, the reprocessing implies direct coupling to the reactor core,...

The objective is to devise the simplest possible system that is compatible with the other constraints. In particular, it will be important to avoid excessive deterioration of the system's fuel regeneration capability.

1.3 Fuel Regeneration Capability

The breeding ratio expresses the balance between the creation of ^{233}U through neutron capture on ^{232}Th and the destruction of ^{233}U through fission or neutron capture. The breeding ratio in a critical reactor can thus be written:

$$BR = \frac{r_{c,^{232}Th} - r_{c,^{233}Pa}}{r_{f,^{233}U} + r_{c,^{233}U}}$$

With r_c and r_f respectively the capture rate and the fission rate of the different isotopes.

A breeding ratio less than 1 implies that ^{233}U is consumed so that fissile matter must be fed into the core on a regular basis. This inevitably increases both the volume and the frequency of transfer of these dangerous materials. Similarly, a breeding ratio larger than 1 implies that the excess ^{233}U produced be placed in storage and/or transported. Because, in all cases, the initial fissile matter inventory has to be produced by other means (e.g. in pressurized water reactors or fast neutron reactors) the highest possible breeding ratio does not necessarily have to be sought.

In order to satisfy the regeneration constraint, we try to achieve a breeding ratio at least equal to 1, knowing that any excess neutrons can always be put to use (improved safety, transmutation capabilities, ...).

[1] The heating of the salt induces a widening of the resonances due to the Doppler effect and a change in the neutron spectrum moderation due to the salt. Both of these effects are considered together, under the term "Doppler".

1.4 Materials Life Span

This concerns in particular how the graphite reacts to irradiation exposure. Beyond a certain degree of damage, it becomes the seat of swelling. Graphite's life span is determined by the time it takes to reach a fluence limit, that we will set to 2.10^{22} n/cm^2 at a temperature of 630 °C [8]. In our calculation, we consider only the neutrons whose energy is larger than 50 keV, i.e. those that create real damage in the graphite.

The goal, with this constraint, is to obtain a life span that is not too short so as to avoid replacing the core graphite too frequently.

1.5 Initial Inventory

The inventory, here, is the amount of ^{233}U needed to start a 1 GWe power reactor. The smaller the inventory, the faster the deployment of a fleet of such reactors can be achieved [9, 10].

Without excluding configurations with a large inventory, its minimization will be sought.

These constraints are not all equivalent; a weighting factor can be assigned to each of them. This factor depends on the technologies available and the goals that guide reactor choices. As the performance of a system depends on how the constraints are weighted and on how difficult it is to satisfy them, it is not possible to specify the "best" solution. The only possibility is to identify a number of interesting trends. This yields a better understanding of the system and can lead to the definition of a power reactor (stringent constraints) or of a demonstration unit (less stringent constraints).

2 Impact of the Parameters on the Constraints

In this section, we examine how various reactor parameters impact the five constraints discussed above. In order to be able to compare the systems studied, we found it useful to define a standard system from which the different studies could stem.

Our standard system is a 1 GWe graphite moderated reactor. Its operating temperature is 630 °C and its thermodynamic efficiency is 40 %. The graphite matrix comprises a lattice of hexagonal elements with 15 cm sides. The total diameter of the matrix is 3.20 m. Its height is also 3.20 m. The density of this nuclear grade graphite is set to 1.86. The salt runs through the middle of each of the elements, in a channel whose radius is 8.5 cm. One third of the 20 m^3 of fuel salt circulates in external circuits and, as a consequence, outside of the neutron flux. A thorium and graphite radial blanket surrounds the core so as to improve the system's regeneration capability. The properties of the blanket are such that it stops approximately 80 % of the neutrons, thus protecting external structures from irradiation while improving regeneration. We assume that the ^{233}U produced in the blanket is extracted within a 6 month period.

The salt used is a binary salt, LiF - $(HN)F_4$, whose $(HN)F_4$ proportion is set at 22 % (eutectic point), corresponding to a melting temperature of 565 °C. The salt density at 630 °C is set at 4.3 with a dilatation coefficient of 10^{-3}/°C [11]. We assume that helium bubbling in the salt circuit is able to extract the gaseous fission products and the noble metals within 30 seconds. The standard reprocessing we consider is the delayed reprocessing of the total salt volume over a 6 month period with external storage of the Pa and complete extraction of the FPs and the TRansUranians (TRU) (Figure 1).

2.1 Influence of the Reprocessing

2.1.1 How Slow Delayed Reprocessing Works

As previously stated, the MSBR reprocessing is considered too complex to be feasible in the next few decades. The effectiveness of this reprocessing rested mainly on the extraction and storage of the protactinium away from the neutron flux so as to avoid, insofar as possible, the production of ^{234}U by neutron capture. The half-life of ^{233}Pa is 27 days and its extraction has to be markedly faster if it is to be efficient. That is why the reprocessing of the total core volume in 10 days was contemplated.

The difficult part of the reprocessing is Fission Product extraction in the presence of thorium. The idea, with slow reprocessing, is to first extract the thorium, so as to avoid being handicapped by its presence in the FP extraction process. This method could not be applied in the MSBR because of the large thorium flow involved, reaching several tons per day while it is only a few hundreds of kilograms per day in the case of a six month reprocessing time.

In addition, with slow reprocessing, the nuclear core can be disconnected from the processing unit, small amounts of the salt being processed individually, instead of resorting to continuous on-line reprocessing, as in the MSBR. This is a source of simplification, it allows easier control of the procedure while making the core less sensitive to possible problems in the reprocessing unit.

Figure 1 gives a general view of what slow reprocessing could entail. Some of the stages shown in this general schematic, such as protactinium storage, can be eliminated while maintaining the primary assets of the reprocessing. Likewise, the neptunium extracted in the course of the first fluorination, and the other TRansUranians can be either reinjected in the core or managed separately. The advantage, in the first option, is that an "incinerating" configuration is obtained, insofar as all the TRUs are kept in the core. With the second option, the production of americium, curium, and other heavier elements is significantly reduced.

The time allocated to cleaning the salt and reinjecting it can be extended considerably. Indeed, if the time needed to reprocess the core volume is equal to the time before reinjecting the salt, there is as much salt outside the core as inside it. Thus, up to 6 months can separate the extraction of the fuel salt and its reinjection in the core, after removal of the FPs. The fissile matter inventory is not increased, however, thanks to the possibility of extracting the uranium during a preliminary fluorination stage. In the case of slow reprocessing, we assume very good extraction efficiencies (they are set to 1 in the calculations) because plenty of time is available.

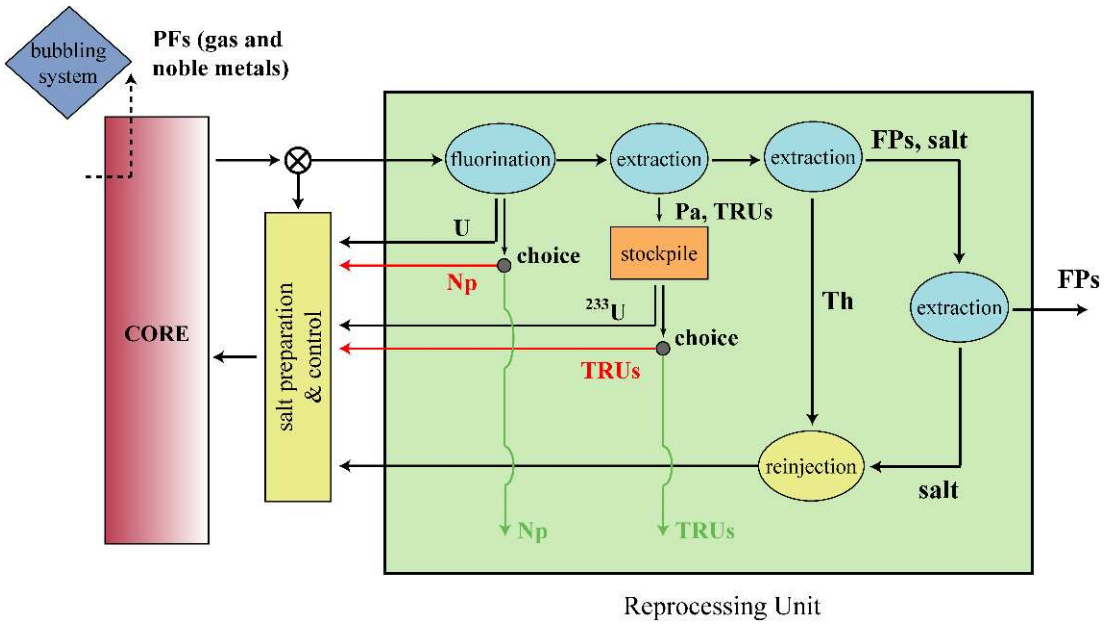

Figure 1: Slow reprocessing overview

It is too early to say that such a reprocessing scheme solves the feasibility issues; it is, however, possible to assert that the simplification of the system improves its feasibility. The impact of this reprocessing on the other constraints, in particular those of regeneration and the feedback coefficients has to be assessed.

2.1.2 Impact of the Reprocessing Time

In Table 1, the breeding ratios obtained at equilibrium are given for various reprocessing options as applied to the reactor configuration described previously. The best breeding ratio is obtained with the MSBR reprocessing and the worst with no reprocessing other than helium bubbling in the core, and ^{233}U recovery in the blanket.

In the table, the MSBR reprocessing is labeled “fast (10 days)” because of the rate at which the protactinium is to be extracted. However, the extraction of the FPs is partial, making the real reprocessing rate longer (equivalent to 50 days for the FPs that capture the most). The option labeled “bubbling only” is set apart because it is dramatically different from the other configurations, making any comparison with them tricky (no equilibrium state).

Varying the reprocessing time from 3 months to 2 years induces about a 0.06 loss in the breeding ratio. For these four configurations, the proportion of protactinium stored outside of the neutron flux is, respectively, 30 %, 20 %, 10 % and 5 %. However, the change in the breeding ratio is due mainly to the change in the capture rate of the FPs and, to a lesser degree, of the TRUs. On the contrary, with fast reprocessing, 80 % of the protactinium is stored outside of the neutron flux and that is the direct cause of the system’s good breeding ratio, way before the FPs and the TRUs. Thus, unless it is extracted rapidly, the Pa’s incidence on regeneration is minor.

We now know the leeway afforded by the reprocessing, since a doubling of the reprocessing time induces a breeding ratio loss of about 0.02. To be precise, we should add that the degradation of the breeding ratio is three times smaller in configurations with a fast neutron spectrum, where the proportion of graphite in the core is reduced.

Reprocessing time	Breeding ratio	dk/dT (pcm/°C)
Fast (10 days)	1.062	-2.25
Slow (3 months)	1.024	-2.37
Slow (6 months)	1.000	-2.36
Slow (1 year)	0.986	-2.39
Slow (2 years)	0.961	-2.50
Bubbling only	0.562	-2.2

Table 1: Breeding ratio and feedback coefficient for several reprocessing options. The statistical error on the feedback coefficient is less than 0.05 pcm/°C.

It is important to note that the reprocessing option chosen has a moderate impact on the feedback coefficients, as shown in the table. This means that reprocessing time and safety can, in a first approximation, be considered to be independent.

2.1.3 Destination of the TRansUranians

As previously stated, the TRUs can either be fed back into the core or they can be managed separately (incinerated in subcritical reactors, incinerated in fast neutron reactors, or placed in storage). The choice has an impact on the regeneration capabilities, as shown in Table 2. Indeed, even if some of the TRUs fission, they impair the neutron balance because of their high capture rates. In the auto-incinerating configuration, the most capturing TRUs reach equilibrium within about 30 years and contribute to the deterioration of the neutron balance.

Reprocessing	Breeding ratio	dk/dT (pcm/°C)
TRUs extracted	1.000	-2.36
TRUs reinjected	0.987	-3.12

Table 2: Breeding ratio and feedback coefficient according to TRU management. The statistical error on the feedback coefficients is less than 0.05 pcm/°C.

In the same table, the influence of the TRUs on the feedback coefficients is also shown. This coefficient is slightly improved if the TRUs are kept in the core. This is because the TRUs harden the spectrum, as will be discussed further in Section 2.2. Note that, in a fast neutron spectrum configuration, the impact of TRU reinjection on both the breeding ratio and the feedback coefficients is reduced.

TRU extraction, however, is advantageous in terms of waste production. When they are submitted to a neutron flux, TRUs form, progressively, significant amounts of very heavy elements such as curium. The ratio of capture to fission cross sections is not favorable to incineration in this type of reactor because of its epithermal neutron spectrum. If these elements are removed from the reactor core, larger amounts of neptunium, formed constantly by captures on ^{236}U, are extracted, but the production rate of the other actinides is reduced, as shown in Table 3. The goal, then, is to obtain TRUs that are more manageable in view of incorporating them in the fuel of Fast Neutron Reactors. If such an outlet for TRUs is not available, this option is of no interest.

	TRUs reinjected (inventory)	TRUs extracted (inventory)	TRUs extracted (output flow)
Np	105 kg	15 kg	4.3 kg / TWh
Pu	265 kg	2.7 kg	770 g / TWh
Am	7.2 kg	0.5 g	0.14 g / TWh
Cm	17.5 kg	0.1 g	30 mg / TWh

Table 3: TRU production and in core inventory at the end of the time period covered by this study (100 years) for two TRU destinations; reprocessing time is 6 months. The output flow calculation is based on 7 TWh per year energy produced.

2.2 Influence of the Size of the Channels

The size of the channels in which the salt circulates is a fundamental parameter of the reactor. Since the size of the hexagons is kept constant in all of our studies, the size of the channels determines the moderation ratio. Changing the radius of the channels modifies the behavior of the core, placing it anywhere between a very thermalized neutron spectrum and a relatively fast spectrum.

The two extreme possibilities correspond respectively to a large number of very small channels and a single big salt channel. In the latter configuration, there is no graphite in the hexagons and the core consists in a single channel. In order to allow a comparison of the results with those of the other configurations, in this case, the hexagons are treated as salt channels with an equivalent area (channel radius: 13.6 cm.).

For the configurations in which the channel radius is equal to or larger than 10 cm, it is essential that the graphite of the axial reflectors be replaced with less moderating materials (e.g. zirconium carbide). Otherwise, the fissions occur massively in the vicinity of the reflectors instead of within the core.

As shown in Figure 2, the radius of the channels has a strong impact on most of the constraints[2].

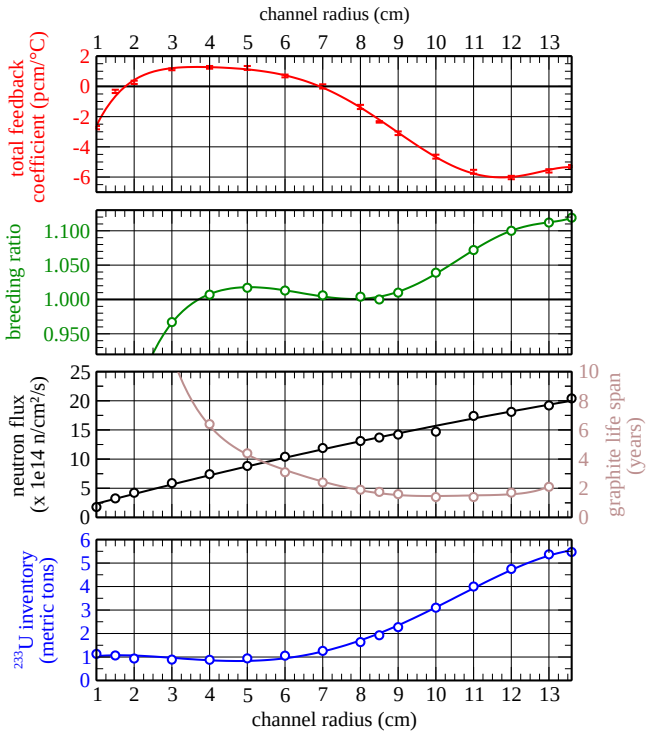

Figure 2: Influence of the channel radius on four of the five constraints (configuration: variable radius, 20 m^3 salt, 630°C, 22% (HN)F$_4$)

2.2.1 Safety Constraint

The study of the feedback coefficients requires a fine analysis of the neutron spectra involved. These are shown in Figure 3 for different channel sizes up to the single channel configuration. The cross section resonances of the materials present in the core have a strong impact on the neutronic behavior of the reactor. The main resonances are visible: fission of ^{233}U at about 2 eV, ^{234}U capture at 5 eV, ^{232}Th capture near 22 eV, and diffusion on ^{19}F near 25, 50 and 100 keV.

As shown in Figure 2, the total feedback coefficient becomes rather strongly negative as the spectrum hardens. This

[2] The moderation ratio can seem to be a more universal parameter but, like the radius of the channels, it is also influenced by other parameters. An identical moderation ratio can yield very different results according to the density of the materials involved or the size of the hexagons.

evolution is due to the conjoined variation of the three sub-coefficients, Doppler, density and graphite, as illustrated in Figure 4.

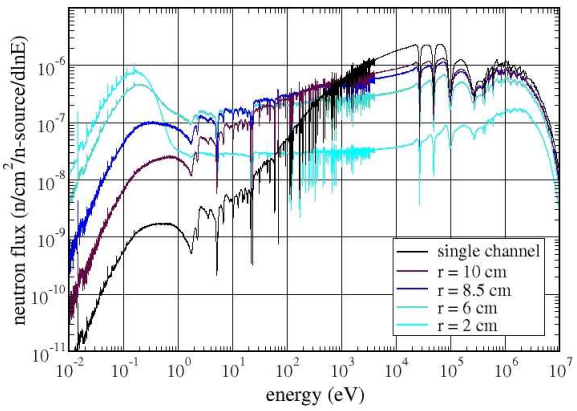

Figure 3: Neutron spectrum for several channel radii (configuration: variable radius, 20 m^3 salt, 630 °C, 22% $(HN)F_4$)

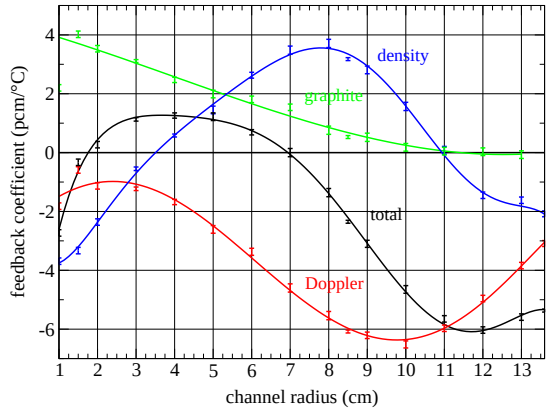

Figure 4: Total feedback coefficient and feedback coefficient components versus channel radius (configuration: variable radius, 20 m^3 salt, 630 °C, 22% $(HN)F_4$)

The Doppler coefficient is linked to the ^{233}U fission resonance and the ^{232}Th capture resonance (and, to a lesser degree, to the ^{234}U capture resonance). These two elements have opposite effects on the feedback coefficient: ^{233}U worsens it whereas ^{232}Th improves it. The thermal agitation of the salt nuclei induces a widening of these resonances so that their influence is increased. The value of the Doppler coefficient depends on how intense the flux is at these resonance values. When the spectrum hardens, the flux is more intense for high energies and less so for low energies, the thorium resonances are favored (main resonance located at 22 eV while that of ^{233}U is at 2 eV). The Doppler coefficient then becomes more negative. Beyond a certain degree of spectrum hardening, the large resonances of both thorium and uranium lie in a zone where the flux has a low intensity and their importance is reduced. This explains the worsening of the Doppler coefficient for large radii.

The density coefficient is related to the expansion of the salt which pushes a fraction of the fuel outside of the moderated zone. The consequence is spectrum softening because the proportion of graphite to salt is larger, thus increasing the fission rate. The effect is small for small radii where thermalization is already very efficient and where it is counterbalanced by captures in the graphite. For large radii, the thermal part of the spectrum contributes practically nothing in the neutron balance and the effects of neutron escape are felt more strongly. The density coefficient can become negative when the effects of captures in the graphite (small radius, large proportion of graphite) or of neutron escapes (large radius, fast neutron spectrum) dominate over the effects of thermalization.

The graphite coefficient comes from an energy shift of the thermal part of the neutron spectrum (around 0.2 eV), due to heating of the moderator. This shift increases the fission rate because of a small low energy (0.3 eV) resonance in the fission cross section of ^{233}U [4]. Its impact on the stability decreases as the amount of graphite in the core decreases and as the influence of the thermal portion of the spectrum weakens.

2.2.2 Regeneration Constraint

The capacity to regenerate the fuel varies a great deal with the size of the channels. This can be explained on one hand by the number of neutrons available and, on the other hand, by the increased neutron losses in configurations with small channel radii. The number of neutrons available represents the number of leftover neutrons once both the chain reaction and the regeneration are ensured. This is defined by:

$$N_a = \nu - 2\,(1+\alpha)$$

Where α is the mean capture to the mean fission cross section ratio of ^{233}U. These available neutrons are distributed mainly between sterile captures and supplementary captures in thorium (breeding). N_a reaches a minimum at about r = 8.5 cm because of the variations of α with the neutron spectrum. For small radii, the strong dip in the breeding ratio is due to neutron losses in the graphite because there is so much of it.

2.2.3 Materials Life Span Constraint

The mean cross sections decrease dramatically with the hardening of the neutron spectrum. As increasing the inventory does not compensate for this loss, the neutron flux has to be increased in order to keep the power constant.

While this phenomenon is linear, this is not true of the core graphite's life span, as shown in Figure 2. A few items have to be stressed:

- The graphite in the center undergoes a flux that is more intense than in the periphery. The life span we provide is averaged over the entire core.
- The maximum fluence in the graphite decreases when the temperature increases and the temperature is not uniform. Since the graphite is heated by gamma radiation

and cooled by the salt, the temperature is higher between channels and lower on the channel surface. The temperature difference increases as the channels are further apart. This means that configurations with a smaller channel radius should have a smaller maximum authorized fluence than those with a larger channel radius.

Generally, the flux in the graphite is directly related to the flux in the salt so that increasing the flux in the salt reduces the graphite's life span. It is thus considerably shorter with larger channel radii than with smaller ones.

As can be noticed, the graphite life span curve does not extend all the way to the single channel configuration since, in that configuration, there is no graphite inside the core (except that of the blanket). This configuration then has an asset in that it almost completely solves the issue of the graphite's life span.

2.2.4 In Core Inventory Constraint

For the reactor to be critical, the fissile matter inventory has to be adjusted when the neutron spectrum hardens. Indeed, the mean ^{233}U fission cross section and the mean ^{232}Th capture cross section decrease as the energy of the neutrons increases but the evolution is not identical for the two isotopes. Two different operating regimes can be singled out as shown in Figure 2.

- For small channel radii, the cross section decreases practically in the same way for the two isotopes and the inventory required does not change much.
- For larger radii, beyond 7 cm, the mean fission cross section of ^{233}U decreases faster than the capture cross section of ^{232}Th so that the inventory has to be increased significantly.

2.3 Influence of the Salt Volume

The power per unit volume of salt (specific power) is a determining parameter in a reactor's behavior. In the reference configuration, it amounts to about 250 W/cm^3 for the salt in the core. This parameter can be modified in two ways: by changing the fuel volume at fixed power or by changing the total reactor power at fixed salt volume. These two options yield similar results and only the first one is discussed in this paper.

Since the flow of Heavy Nuclei is considered to be a key factor for the feasibility of the chemical reprocessing, the reprocessing time is adjusted so as to keep this flow constant from one system to the other. Thus, doubling the salt volume implies that core reprocessing takes twice as long. The incidence of the salt volume on the various constraints is rather simple, as shown in Figure 5.

The size does not have a significant impact on the feedback coefficients because the neutron spectrum changes very little with the size. The slight evolution of the coefficient is due to the difference in neutron escapes, which are more likely in smaller reactors.

The evolution of the breeding ratio as the salt volume increases has two main causes: the difference in neutron escapes, and the change in specific power, losses due to the Pa being

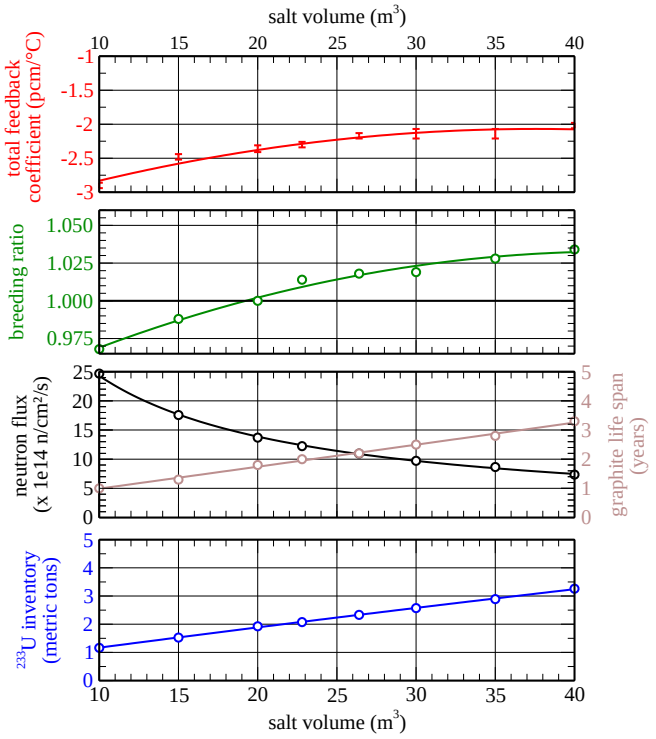

Figure 5: Influence of the salt volume on four of the five constraints (configuration: r = 8.5 cm, variable salt volume, 630°C, 22% (HN)F_4)

a direct function of the specific power. FP capture rates do not play a significant role in this evolution. Unlike Pa, whose inventory is determined mainly by its rapid radioactive decay, their concentration at equilibrium depends on the reprocessing time. The longer reprocessing time exactly compensates the effect due to the smaller specific power.

The neutron flux in the core is directly related to the specific power and the graphite's life span varies accordingly. Similarly, the inventory in the core depends on the volume but the effect is not directly proportional, because neutron escapes are different.

One last important aspect is the thermal hydraulic constraint. The thermal power is evacuated by the fuel which thus has to circulate in the exchangers. A limit has to be set on the out of core salt volume so as to allow reactor control: the delayed neutrons precursors migrate away from the neutron flux along with the salt. In our studies, the external salt volume is 1/3 of the total volume. Heat evacuation becomes more difficult as the specific power increases. Small sized or high power reactors are at a disadvantage in this respect.

2.4 Influence of the Salt Composition

2.4.1 Elimination of the Be

By definition, the salt plays a central role in MSRs. Serving as the solvent for the fuel, as the moderator and as the coolant, it has to have many characteristics specific to the neutronic as well as the chemical, hydraulic, or thermal aspects. Like the

MSRE, the MSBR was based on a fluoride salt, because of its good neutronic properties (capture rate and moderating capability) in a thermal spectrum. Lithium was chosen for the same reasons and beryllium because it brought the melting temperature down to 490 °C. The salt composition was 71.7% LiF - 16% BeF_2 - 12.3% $(HN)F_4$ [3].

Our first step in studying the influence of the salt composition was to eliminate beryllium from the salt bringing it to the eutectic point 78% LiF - 22% $(HN)F_4$. All the studies discussed up to now were done with this composition. The reasons for eliminating beryllium are based mainly on problems with its chemistry, its toxicity, and its availability. The proportion of heavy nuclei in the eutectic changes drastically and its melting point increases from 490 °C to 565 °C [4]. This temperature increase remains moderate and it seems manageable with the commonly used structure materials. Because the significantly higher proportion of Heavy Nuclei has a strong impact on the in core inventory and on the breeding ratio, we have decided to reduce the salt volume from 40 m^3 (MSBR) to 20 m^3 (reference configuration for these studies) so as to keep the same amount of Heavy Nuclei in the reactor and, as a result, similar neutronic behavior.

The elimination of the beryllium impacts all five constraints. The presence of a ternary component in the salt seems to complicate the reprocessing chemistry, the risk being that this element be extracted instead of the target elements. Thus, using the LiF - $(HN)F_4$ salt could simplify fuel reprocessing and, as a result, bring it closer to feasibility. The impact on the other constraints is shown in Table 4. The spectrum is harder with the binary salt because of the larger proportion of HN. This translates directly into an improvement of the feedback coefficient and a larger inventory. The change in the breeding ratio is due jointly to the increased proportion of HN (positive action) and to the increased specific power (negative action). The latter also implies a neutron flux increase which, combined with the faster neutron spectrum, leads to a significant deterioration of the graphite's life span.

	LiF - BeF_2 (40 m^3)	LiF (20 m^3)
Feedback coeff. (pcm/°C)	-1.57	-2.36
Breeding ratio	1.009	1.000
Neutron flux ($x10^{14}$ n/cm^2/s)	8.6	13.7
Graphite life span (years)	3.3	1.8
^{233}U inventory (kg)	1650	1925

Table 4: Constraints according to the type of salt used to dissolve the fuel. The statistical error on the feedback coefficients is less than 0.05 pcm/°C (configuration: r = 8.5 cm, variable salt volume, 630 °C, variable $(HN)F_4$ proportion).

The elimination of beryllium has an additional and significant advantage, that is not related to the constraints that we have identified in this paper. Tritium is produced, in a system like the MSBR, by the (n,nt) reaction on ^7Li and the (n,t) reaction on ^6Li, producing 2/3 and 1/3 respectively of the tritium [4]. The lithium used is 99.995 % enriched with ^7Li; the ^6Li is rapidly consumed, unless it is regenerated by an (n,α) reaction on ^9Be. With the elimination of beryllium, this reaction cannot occur and, as a result, the production of tritium is reduced.

2.4.2 Evaluation of the LiF - $(HN)F_4$

Temperature increase : The proportion of Heavy Nuclei in the binary salt can be adjusted. As it is reduced, the melting point increases, reaching 845 °C with pure LiF. Common structure materials cannot withstand such a temperature increase. However, new promising solutions based on carbon (carbon-carbon, carbon fiber, carbides,...) could help solve this problem [12]. If this technology can be implemented, then the HN proportion parameter can be modified.

	630°C	1030°C
Feedback coeff. (pcm/°C)	-2.36	-1.00
Breeding ratio	1.000	1.026
Neutron flux ($x10^{14}$ n/cm^2/s)	13.7	9.6
Graphite life span (years)	1.8	1.2
^{233}U inventory (kg)	1925	1630

Table 5: Constraints according to the mean temperature of the fuel salt. The statistical error on the feedback coefficients is less than 0.05 pcm/°C (configuration: r = 8.5 cm, 20 m^3 salt, variable temperature, 22% $(HN)F_4$)

The temperature increase due to the change of salt leads us to set the operating temperature at 1030 °C for all the configurations. We will first study the influence of this temperature hike on the standard configuration before studying the influence of the HN proportion in the salt at 1030 °C.

At this temperature, the thermodynamic efficiency is assumed to increase from 40 % to 60 % and this has an incidence on the thermal power of the reactor: 1666 MWth instead of 2500 MWth are needed to produce 1000 MWe. Similarly, the salt density decreases form 4.3 to 3.89 because of the temperature related expansion effect. The impact on the constraints of this temperature increase is detailed in Table 5.

The change in salt density has a direct influence on the moderation ratio, resulting in a better thermalization of the neutron spectrum. This induces, for a channel radius of 8.5 cm, a worsening of the feedback coefficient (a behavior similar to that shown in Figure 4). Similarly, this slight thermalization leads to a larger ^{233}U fission cross section and, combined with the lower salt density, a smaller necessary inventory. As for the breeding ratio, it is improved because of the reduced specific power, which has a direct incidence on parasitic captures (mainly those of FPs and Pa). The lower specific power has an incidence also on the neutron flux and, thus, on the graphite's life span. However, at such a temperature, the fluence limit that the graphite can withstand is reduced from 2.10^{22} n/cm^2 to

[3] The composition that we really used in our tests with this type of salt [4] is: 70% LiF - 17.5% BeF_2 - 12.5% $(HN)F_4$.

[4] In order to make comparisons easier and because the temperature difference is not large, the studies with the 78% LiF - 22% $(HN)F_4$ salt were done at the same temperature as in previous studies [3, 4], i.e. 630 °C.

10^{22} n/cm^2 [8]. As a result, the graphite's life span is reduced in spite of the smaller neutron flux.

Influence of the proportion of Heavy Nuclei: Now that the effect of the temperature change from 630 °C to 1030 °C is known, the impact of the proportion of Heavy Nuclei can be explored. It is useful to keep the total amount of Heavy Nuclei constant, as we did when we changed the salt composition. As a consequence, salts with a smaller proportion of HN will have a larger volume. This makes these new configurations potentially interesting from the point of view of thermal power extraction. The core reprocessing time is kept at 6 months since, in that case, the flow of HN to be reprocessed is the same for all the configurations. The salt of the thorium blanket is not modified. The density and expansion coefficient of the fuel salt are crucial parameters, they are given in Table 6 [11].

Since the graphite's life span is directly related to the specific power and since the inventory is kept constant, these constraints are not very interesting in this part of our study. We will thus concentrate our attention on the safety and the regeneration constraints. Rather than presenting the impact of the proportion of HN for a reference configuration (r = 8.5 cm, salt volume = 20 m^3) as was done previously, we will look at the impact of the channel radii for different HN proportions. This view point will allow a better understanding of the phenomena at play. The results are shown in Figures 6 and 7.

	22%	10%	5%	2%
Salt volume (m^3)	20	36.8	67.2	155
Density	3.89	2.85	2.33	1.98
Expansion coeff. (x10^{-4}/°C)	10	10	9	8

Table 6: System properties according to the percentage of $(HN)F_4$

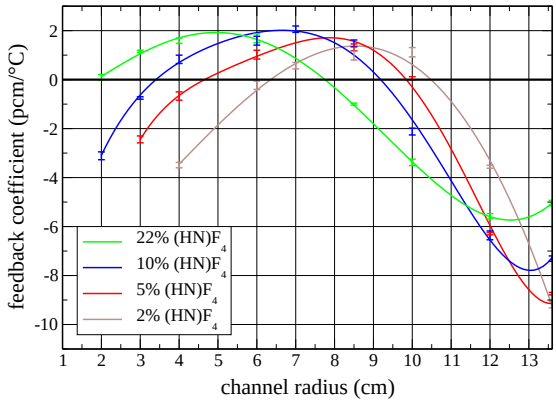

Figure 6: Feedback coefficient versus channel radius for several proportions of $(HN)F_4$ (configuration: variable radius, variable salt volume, 1030 °C, variable proportion of $(HN)F_4$).

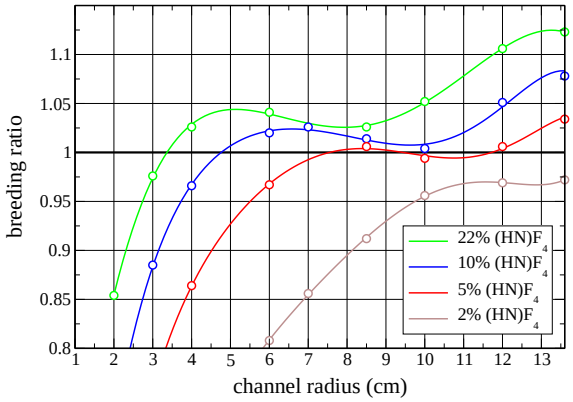

Figure 7: Breeding ratio versus channel radius for several proportions of $(HN)F_4$ (configuration: variable radius, variable salt volume, 1030 °C, variable proportion of $(HN)F_4$).

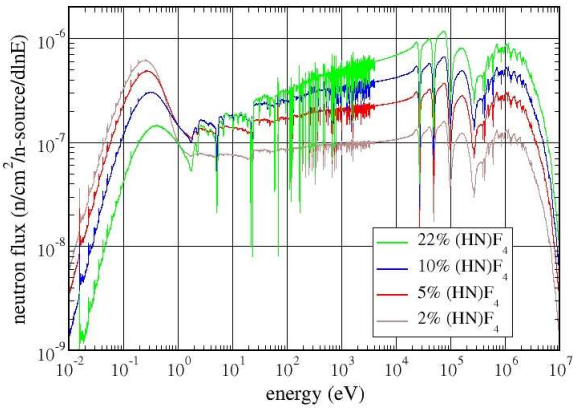

Figure 8: Neutron spectra for several proportions of $(HN)F_4$ (configuration: r = 8.5 cm, variable salt volume, 1030 °C, variable proportion of $(HN)F_4$).

During the reprocessing outlined in Figure 1, the thorium is extracted in order to allow the extraction of the FPs from the salt. This step is a key point in the reprocessing and it is made easier if the proportion of HN in the salt is small.

Moreover, when this proportion is decreased, the neutrons are scattered for a longer time before they encounter a fissile or fertile element (those that dominate neutron absorptions). This leads directly to a more thermalized neutron spectrum, as shown on Figure 8. Thus, the behavior of configurations with a small proportion of HN is similar to that of configurations with a 22% $(HN)F_4$ proportion but with smaller channels.

This additional thermalization is the main cause of the evolution of the feedback coefficients. And the visible difference for the configurations with a single salt channel seems to be due mainly to the improvement of the Doppler sub-coefficient. Likewise, the breeding ratio curves are similar from one salt to the other but two effects are observed. The first effect is related

to the thermalization change and the second to a deterioration of the breeding ratio when the HN proportion is decreased. This is due to an increased capture rate in the light elements of the salt on which the neutrons scatter for a longer time.

3 General Discussion

The various studies that have been carried out lead to a better understanding of the way an MSR works. The search for reactor configurations, be it a demonstrator or a power generator, requires that a certain number of constraints be satisfied. To do so, the different studies discussed in this paper have to be combined and the results extrapolated. Since the parameters are not mutually independent, one has to be circumspect in this approach.

Let us try to explore the possible reactor configurations. For the sake of simplicity, the parameter concerning the salt composition will not be considered in a first approach. The salt, then, is 78% LiF - 22% $(HN)F_4$ and the mean temperature is 630 °C.

Since the safety aspect cannot be circumvented in the design process of a nuclear reactor, we consider that this constraint is necessarily satisfied. Moreover, we consider only those configurations whose total feedback coefficient, not just the salt feedback coefficient, is negative. Except in the case where the size of the reactor is reduced dramatically, leading to a significantly increased neutron flux, the total feedback coefficient is negative only for either very thermalized or fast neutron spectra.

The first option implies a small fissile matter inventory and a weak neutron flux. When submitted to such a flux, the graphite undergoes little damage and its life span is reasonably long. On the other hand, captures in the moderator deteriorate the breeding ratio significantly. If a reactor system does not need to regenerate its fuel, then this very thermalized configuration may be suitable.

The faster neutron spectrum option introduces a real difficulty concerning the graphite, whose life span is then on the order of one year. There are several solutions to this problem.

- Decreasing the specific power of the reactor (by increasing its size and/or decreasing the total power generated) leads directly to a decreased flux intensity and, as a consequence, extends the graphite's life span. This, however, increases in the same proportion the per GWe fissile matter inventory, without providing a very satisfactory solution.
- The absence of moderating graphite in the single salt channel configuration solves this problem, the graphite in the periphery of the core being much less irradiated. However, this option leads to the fastest neutron spectrum and, as a result, the largest fissile matter inventory (5 to 6 metric tons of ^{233}U). The specific power can be increased, though, in order to reduce the per GWe inventory.
- It may be possible to use a material whose structure is much less sensitive to irradiation than graphite. Then the configuration space for channel radii lying between 6 and 13 cm would no longer be forbidden.

Finally, the high breeding ratio obtained thanks to the fast neutron spectrum provides more leeway for the reprocessing. In particular, the time for full core reprocessing can be significantly extended. The single salt channel configuration discussed earlier can even do without any reprocessing (except for the bubbling process and uranium retrieval in the blanket) and still regenerate its fuel during the first 20 years of operation. The salt could conceivably be replaced, or entirely reprocessed after this time has elapsed. If a 6 month reprocessing time is kept, it becomes possible to do without a thorium blanket while still regenerating the fuel. This would substantially simplify the reactor design. Finally, we should note that, with a fast spectrum, re-injecting the TranUranians in the core would be interesting, both in terms of regeneration (small neutron losses) and in terms of waste production (good incineration capability).

If it proves possible, increasing the temperature to above 1000 °C has many positive repercussions on the constraints, in particular thanks to the increased thermodynamic efficiency. However, it induces better thermalization by the salt and this has to be taken into account.

This opens the way to salt compositions containing small amounts of Heavy Nuclei. The way these reactors behave is practically the same with such compositions as with 22% $(HN)F_4$. In particular, the neutron spectra have to be either very thermalized or fast to ensure negative feedback coefficients. However, this corresponds to different reactor configurations because of the increased thermalization due to the salt's light nuclei. The capture rate of these light elements deteriorates the breeding ratio and leaves less leeway than with compositions containing more HN. The fundamental importance of this parameter lies elsewhere. Indeed, it is possible either to keep the size constant, thus reducing the inventory but not the specific power, or to increase the size, thus decreasing the specific power but not the inventory. A compromise between these two extremes can be found since decreasing the specific power facilitates the evacuation of the thermal power, a constraint that must not be neglected.

Conclusion

While our studies were, at first, close to the MSBR configuration, they prompted us to diversify our investigations. We analyzed the impact on the behavior of the core of such parameters as the reprocessing, the moderation ratio, the core size, and the proportion of heavy nuclei in the salt.

Our results confirm that there is a problem with the feedback coefficients in the MSBR. In a thermal spectrum, it would be possible to reach an acceptable concept only after in depth investigations taking into account the effect of the salt (negative feedback coefficient) and of the graphite (which makes the global feedback coefficient positive) separately. For very thermalized spectra, the global coefficients are negative thanks to the large neutron losses in the moderator but this leads also to a very poor breeding ratio. Epithermal or fast neutron spectra thus seem more favorable since they combine good feedback coefficients with satisfactory breeding ratio. However they lead to severe problems with the graphite's ability to withstand the

irradiation. As a result, the solution that removes the moderating block seems especially attractive.

Our studies have uncovered a wider range of possibilities than anticipated. Thus, many options remain to be explored. In particular, the evaluation of new materials, be it to obtain a moderator that has better irradiation resistance properties or to allow high temperature operation is crucial for an even more interesting development of the concept. How to extract the thermal power from the core is another issue of major interest since it impacts the behavior of the core through the specific power aspect. In order to ease heat recovery, the salt composition can also be modified so as to dissolve the fissile matter in a larger salt volume. In general, it is possible to change the type of salt, the MSR concept being adaptable to such a change.

As many parameters remain to be studied, other acceptable solutions could be found. In particular, parameters such as the type of salt, the moderating material, the size of the lattice hexagons, the definition of several different areas in the core, could be studied more specifically. In view of the results already obtained, it is clear that many configurations remain to be explored, requiring research on the salt and the materials as well as on the neutronics and the geometry.

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